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(54) Title: PLASTIC FILM HAVING IMPROVED IMAGING PROPERTIES

(57) Abstract: The multilayer plastic films of the invention of three or more layers are formed from a polypropylene core layer, a top skin layer formed from polypropylene or a polypropylene copolymer, and a bottom skin layer of polypropylene. The optical properties of the films are enhanced for imaging and labeling uses by the inclusion of one or more colored tints in the top skin layer or in the top tie layer. The core layer may contain a cavitating agent and the bottom skin layer may contain an antiblocking additive. Optionally, the films may also include a top tie layer formed of polypropylene and located between the top skin layer and the core layer. The top tie layer may include titanium dioxide and may further include an optical brightener. The film may also include a bottom tie layer formed of polypropylene and located between the core layer and the bottom skin layer.

PLASTIC FILM HAVING IMPROVED IMAGING PROPERTIES

BACKGROUND OF THE INVENTION

The present invention relates to the field of complex multilayer plastic films of polyolefin compositions suitable for use in packaging and labeling. In 5 particular, the present invention relates to improved films with enhanced optical properties incorporating colored tint components.

Clear and opaque multilayer plastic films are used in packaging for 10 applications where images or designs are printed or applied to the film surface. The colors of these images and designs on the currently available clear or translucent plastic labeling films are stark, and generally lack any illusion of depth. The images and designs appear to be flat and limited to two-dimensional representations.

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Currently available opaque plastic films also have undesirable imaging properties. In particular, the visual quality of affixed or printed colored images and designs on opaque films, like those of clear films, are stark and lacking in depth. Moreover, their suitability for packaging and labeling applications is even 20 more limited due to the lack of transmitted light to illuminate any images or designs they may carry.

The marketing and customer appeal of these currently available films is limited by the perceived flatness of the images and designs applied to the film 25 surface, and a starkness of their incorporated colors.

There is a therefore need for labeling and packaging films that support printed or affixed colorful images and designs with more warmth, and exhibiting more depth of color. The images should also have greater clarity and 30 brightness.

SUMMARY OF THE INVENTION

The present invention provides plastic films of three, four or five layers. The films have enhanced optical properties and are useful for imaging applications.

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The three layer films have the following structure: A core layer which is formed from polypropylene and may include a cavitating agent; a top skin layer, composed of a polyolefin polymer, one surface of which forms the top exposed surface of the film; and a bottom skin layer one surface of which forms the 10 bottom exposed surface of the film. The bottom skin layer is formed from polypropylene, polyethylene or a polypropylene copolymer. The polyolefin polymer of the top skin layer is formed from a propylene-ethylene copolymer, a high-density polyethylene polymer, a low-density polyethylene polymer, or 15 polypropylene. At least one image enhancing colored tint is included in the top skin layer.

Alternatively, the films of the present invention may be formed from four layers: A core layer formed from polypropylene and which may include a cavitating agent; a top skin layer formed from a polyolefin polymer, one surface 20 of which forms the top exposed surface of the film; a top tie layer formed from polypropylene, being located between the top skin layer and the core layer; and a bottom skin layer formed from polypropylene, polyethylene or a polypropylene copolymer; one surface of which forms the bottom exposed surface of the film. The polyolefin polymer of the top skin layer is formed from a propylene 25 copolymer, a high-density polyethylene polymer, a low-density polyethylene polymer, or from polypropylene. One or more image enhancing colored tints may be included in the top skin layer or in the top tie layer. Alternatively, the top skin layer and the top tie layer may each include one or more image enhancing tints. The top tie layer may contain TiO₂ as a whitening agent and may further include 30 an optical brightener.

Finally, the films of the present invention may be formed from five layers: a polypropylene core layer; a top skin layer formed from a polyolefin; a bottom skin layer formed from polypropylene, polyethylene or a polypropylene copolymer; a top tie layer formed from polypropylene and located between the 5 top skin layer and the core layer; a bottom tie layer formed from polypropylene located between the core layer and the bottom skin layer. The polyolefin polymer of the top skin layer is formed from a propylene-ethylene copolymer, a high-density polyethylene polymer, a low-density polyethylene polymer, or from polypropylene. One or more image enhancing colored tints are included either in 10 the top skin layer, or in the top tie layer. In another embodiment, one or more colored tints are included in the top skin layer and further, one or more image enhancing colored tints are included in the top tie layer. Each colored tint may be distributed in one or both layers. The top tie layer may also contain TiO_2 as a whitening agent and may further include an optical brightener.

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DETAILED DESCRIPTION OF THE INVENTION

The plastic films of the invention may be formed from three or more polyolefin layers and may be clear, opaque, translucent or white depending on the nature and amounts of additives included in the layers. The films are stretched 20 multilayer films with a core layer of a polyolefin, preferably isotactic, biaxially oriented polypropylene. The optical characteristics of the films, especially the brightness and depth, or warmth of the colors of applied images are determined by the combination of tints, whiteners and brighteners included in the various layers of the films. The colored tints enhance the optical properties of any images or 25 designs applied onto the film surface. Preferably, the image or design is printed or applied on to the top skin layer.

The films of the present invention comprise at least three layers: a core layer which may include a cavitating agent, and two outer skin layers, the top skin 30 layer and the bottom skin layer. The top skin layer being juxtaposed to one surface of the core layer and the bottom skin layer being juxtaposed to the other

surface of the core layer. The core layer is preferably formed from isotactic, biaxially oriented polypropylene.

One of the outer skin layers of the films of the invention, herein designated 5 as the top skin layer, is formed from a polyolefin. The top skin layer may include one or more colored tint compounds. The second outer skin layer is herein referred to as the bottom skin layer and is formed from polypropylene or a polypropylene copolymer. This bottom skin layer may also include an antiblocking additive for improved machining properties.

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Polyolefins useful in forming the top skin layer include polyolefin terpolymers, polyolefin copolymers and polyolefin homopolymers. Examples of polyolefin polymers that are particularly useful for forming the top skin layer include propylene-ethylene-butylene terpolymers, propylene-ethylene copolymers, 15 high-density polyethylene polymers, medium density polyethylene polymers, low density polyethylene polymers, and polypropylene or blends of any of the above polyolefins.

The bottom skin layer may be formed from polypropylene, polyethylene or 20 a polypropylene copolymer. Preferred polypropylene copolymers for forming this layer include polypropylene-ethylene copolymers, with polypropylene-ethylene copolymers containing less than about 10% by weight of ethylene being more preferred, and about 5% ethylene being optimum. Alternatively, the bottom skin layer may include a blend of polypropylene and polyethylene.

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When propylene-ethylene-butylene terpolymers, ethylene-propylene copolymers or propylene-ethylene copolymers are incorporated into an outer layer of the film, heat-sealing properties are imparted to the film.

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The three layer films of the present invention are preferred for certain applications requiring clear films for receiving colored images or designs. The images or designs are printed or affixed to the exposed surface of the top skin

layer, which may be treated to improve receptivity to printing inks or adherence of affixed images or designs as described below.

The four-layer films of the invention include a polypropylene core layer; a 5 top tie layer also formed from polypropylene and located on one surface of the core layer designated as the top surface; a polyolefin top skin layer located adjacent to the top tie layer forming the top surface of the film; and a bottom skin layer formed from polypropylene or a polypropylene copolymer and located adjacent to the second surface of the core layer. The four layer films include one 10 or more colored tint compounds in the top skin layer or in the top tie layer, or both. The top tie layer may also include a whitening agent and may further include an optical brightener. The four layer films are preferred for certain applications including transparent films, translucent films and white films.

15 The five layer films of the invention are optimal for machining in production, printing or labeling and packaging. The five layer films have certain advantages over the three layer films. For example, three layer films with abrasive additives such as TiO₂ included in a surface layer are prone to plate out: the deposition of additive on machinery surfaces, causing surface deterioration 20 and accelerated wear of costly machinery.

The five layer films of the invention also have advantages over four layer films including providing two separated tie layers for inclusion of incompatible components and better machining properties. The latter include avoidance of 25 stress distortions, curl and rippling, as well as improved surface properties. These improved surface properties, include better smoothness and reduced marring due to protrusions caused by the cavitating agent in the core layer.

In one aspect of the invention the colored tint, incorporated into one or 30 both of the top skin layer and the top tie layer, is a red tint. The red tint may be incorporated into the films of the invention to improve the visual quality of an image printed or affixed to the top skin layer. The red tint may be any red tint that

provides improved flesh tones and rosy appearance or apparent warmth of any image printed or applied to a surface of the film.

For the imaging applications of the present invention a white plastic film 5 with slight pink tinge is preferred. In a preferred aspect the red tint is an acridone tint, such as for instance a quinacridone or a derivative of a quinacridone. An example of a particularly preferred red tint useful in enhancing the optical properties of the films of the present invention is the acridone tint, RT-790 produced by Ciba-Geigy.

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In another aspect the colored tint is a blue tint. The blue tints enhance the visual whiteness of the film. For marketing appeal a white plastic film with slight bluish tinge is preferred. The blue tints useful for providing enhanced optical properties in the films of the invention include blue dyes and blue pigments. Blue 15 dyes may suffer from photo-bleaching, whereas blue pigments are more stable to light, and are preferred. Blue pigments useful in the films of the present invention include phthalocyanine blue pigments, cromophtal blue pigments, irgazin blue pigments and irgalite organic blue pigments. Other blue pigments that may be incorporated into the film layers include cobalt aluminate pigments and cobalt 20 chromium aluminate inorganic blue pigments. An example of a particularly preferred blue tint is the Cobalt Blue tint, Sheppard Blue 214 produced by Sheppard Chemical Company.

Addition of colored tint up to about 0.2% by weight of the total film is 25 contemplated, with about 0.005% to about 0.15% being preferred, and about 0.007% to about 0.1% being optimal. In any particular layer of the film, the colored tint should preferably not exceed 2% by weight of the layer.

Polypropylene is the preferred polyolefin composition for the core layer 30 because of its excellent physical properties and suitability for multilayer thin film manufacturing. These properties include high strength, suitable cavitation properties for production of low-density films, low melting temperature and good

surface properties. Other advantages of polypropylene over other polyolefins include its low cost and wide availability.

The density of the films of the invention may be decreased by inclusion of 5 particles of a cavitating agent such as polybutylene terephthalate (PBT) in the core layer. The particles of the cavitating agent form voids in the softer matrix of the core layer during biaxial stretching of the film. Increasing the content of the particulate cavitating agent in the core layer increases the abundance of 10 microvoids in the stretched film, contributing to lowering the overall density of the film. This biaxial stretching orients the polymer molecules along the perpendicular stretched axes. Biaxial orientation imparts other desirable physical characteristics to the film, including increased tear strength and heat shrinkage properties.

15 Up to about 25% by weight of a cavitating agent may be incorporated into the core layer of films of the invention. More preferably, up to about 15% and optimally up to about 10% of the cavitating agent may be included in this layer. For translucent films, up to about 6% of the cavitating agent may be included in the core layer. Preferably, up to about 5%, and optimally about 4% of the 20 cavitating agent such as PBT is included in the core layer.

Opacity may be further enhanced by the addition of whiteners or opacifying agents to one or more layers of the film. These include TiO_2 , $CaCO_3$, $BaSO_4$, ZnS , $MgCO_3$, clay, talc, kaolin or any other highly reflective white 25 compound. TiO_2 is a particularly favored whitener for use in the films of the present invention. Preferably, neither whiteners nor opacifying agents are included in any layer that contains a colored tint. Furthermore, any whiteners or opacifying agents should not be located in a layer between the image or design and the layer(s) that contain a colored tint. This prevents any interference with the 30 image enhancing properties of the tint(s) due to the intervening whiteners or opacifying agents.

Up to about 20% by weight of TiO₂ may be included in the top tie layer to enhance whiteness of the film and reflect light through a layer containing a colored tint to brighten any affixed or printed image applied to the film. Inclusion of up to 10% TiO₂ is preferred. Optimally from about 4% to about 6% of TiO₂ 5 may be incorporated into the top tie layer of opaque and white films. For clear films the TiO₂ may be omitted.

Other additives that enhance the optical properties of the film include 10 optical brighteners. Optical brighteners are typically fluorescent agents that absorb energy in the ultraviolet region and emit light largely in the blue region. The following examples are intended as illustrations only, and are not to be taken as limiting in any way: Optical brighteners which may be incorporated into one or 15 more layers of the films of the present invention include 4,4'-diaminostilbene-2,2'-disulfonic acid, derivatives of 4,4'-diamino-stilbene-2,2'-disulfonic acid, coumarin derivatives such as 4-methyl-7-diethylamino-coumarin, 1,4-bis(O-cyanostyryl)benzol, 2-amino-4-methyl-phenol, and 2,2'-(1,2-ethene-diyl)-4,1-phenylene) benzoxazole. Examples of preferred optical brighteners include Eastobrite® OB-1 produced by Eastman Chemical Company, USA and Horstalux® produced by BASF.

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The inclusion of up to about 2% by weight of an optical brightener in any one of the layers of the film is contemplated in the present invention. Preferably from about 0.05% to about 0.5% optical brightener is included in one layer; and optimally about 0.2% optical brightener is included in a single layer, most 25 preferably the top tie layer.

Other additives that may be usefully incorporated into one or more layers of the films include antistatic agents, anti-condensing agents and anti-oxidants. Examples of suitable antistatic agents include such compounds as cocoamine, 30 N,N-bis(2-hydroxyethyl)sterylamine, or any of a variety of monoamines, diamines and tertiary amines well known in the art. The anti-condensing agent may be any

anti-condensing agent such as for instance a fluoropolymer. The anti-oxidant may be any anti-oxidant, such as for example a phosphite.

Further useful additives that may be incorporated into one or both surface 5 layers of the films of the present invention include antiblock agents and slip agents. These additives are incorporated into the films to prevent sticking and to reduce the coefficient of friction, respectively. These properties improve the characteristics of the films for ease of manufacturing and processing, particularly in modern high-speed machinery for rolling and packing.

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Antiblock agents and slip agents include various forms of coated or uncoated silica, silicones, siloxanes, silicon oils and cross-linked silicones. Other useful anti-block/slip agents also include for example methyl acrylate and the non-migratory slip agents as well as many others well known in the art. Particularly 15 useful non-migratory slip agents include ethylene vinyl alcohol (EVOH) and ethylene vinyl acetate (EVA). Preferably the antiblock is included in a skin layer or applied onto the exposed surface of a skin layer. Optimally the anti-block/slip agent is included in the bottom skin layer, or applied onto the exposed surface of the bottom skin layer.

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Up to about 1% of an antiblock agent or slip agent may be included in a skin layer of the films of the invention. More preferably, from about 0.1% to about 0.5% of the antiblock agent or slip agent, and optimally, from about 0.15% to about 0.25% of the antiblock agent or slip agent may be included in the skin 25 layer. Most preferably the antiblock agent or slip agent is included in one skin layer and the colored tint(s) are included in the other skin layer.

The films of the present invention may be treated on one or both of the exposed surfaces of the top skin layer to improve the functionality of the 30 surface(s), including for example, receptivity for printed images and designs, particularly for water-based inks, or for adherence of affixed label. Such surface treatments may include corona treatment, flame treatment, metalizing treatment or

vacuum deposition and other surface treatment methods well known in the art. Treatment of the exposed outer surface of the top skin layer is preferred.

The films of the invention are particularly useful for receiving digital
5 images, which may be applied from an ink jet printing or thermal transfer process. Alternatively, the films may receive images transferred from a master printing plate or applied via an intermediate substrate. Such methods are especially useful for particularly demanding individualized printing applications, such as for instance, for bar coding or individual numbering of packaged or labeled items.

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With the red and blue tints and an optical brightener added into the film, the printed image on the surface shows an intensified clarity and brightness. In addition, the image resolution is greatly improved. These films are suitable for packaging and label applications where high image quality is desired.

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The films of the present invention may be of any thickness, although films with a total polymer gauge thickness from about 0.3 mil to about 5 mil are preferred. More preferably the films have a polymer gauge thickness from about 0.5 mil to about 3 mil. Optimally, the polymer gauge thickness of the films is in
20 the range from about 1 mil to about 2 mil. However, in films that include a cavitating agent cavitation may increase the actual optical gauge thickness by 50% or more.

The three layer films of the invention have a top skin layer with a polymer
25 gauge thickness from about 0.02 mil to about 0.1 mil; a bottom skin layer with a polymer gauge thickness from about 0.05 mil to about 0.5 mil; and a core layer with a polymer gauge thickness such as required to bring the total film polymer gauge thickness to between about 0.5 mil and about 3 mil.

30 The preferred three layer films have a top skin layer with a polymer gauge thickness from about 0.03 mil to about 0.05 mil; a bottom skin layer with a polymer gauge thickness from about 0.1 mil to about 0.4 mil; and a core layer

with a polymer gauge thickness such as required to bring the total film polymer gauge thickness to between about 1 mil and about 2 mil.

In another embodiment, the film of the present invention comprises four
5 layers: a top skin layer having a thickness from about 0.02 mil to about 0.1 mil; a top tie layer having a thickness from about 0.05 mil to 0.5 mil; a bottom skin layer having a thickness from about 0.05 mil to about 0.5 mil; and a core layer having such thickness as required to bring the polymer gauge of the film to a thickness from about 1 mil to about 3 mil.

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Preferably, the layers of the four layer film each have polymer gauge thickness as follows: the top skin layer has a thickness from about 0.03 mil to about 0.05 mil; the top tie layer has a thickness from about 0.1 mil to 0.4 mil; the bottom skin layer has a thickness from about 0.1 mil to about 0.4 mil; and the
15 core layer has such thickness as required to bring the polymer gauge thickness of the entire film to a thickness from about 1.5 mil to about 2.5 mil.

In yet another embodiment, the film of the present invention comprises five layers: a top skin layer having a thickness from about 0.02 mil to about 0.1
20 mil; a top tie layer having a thickness from about 0.05 mil to 0.5 mil; a bottom tie layer having a thickness from about 0.05 mil to about 0.5 mil; a bottom skin layer having a thickness from about 0.02 mil to about 0.1 mil; and a core layer having such thickness as required to bring the polymer gauge thickness of the entire film to a thickness from about 1 mil to about 3 mil.

25

Preferably, the layers of the five layer films each have polymer gauge thickness as follows: the top skin layer has a thickness from about 0.03 mil to about 0.05 mil; the top tie layer has a thickness from about 0.1 mil to 0.4 mil; the bottom skin layer has a thickness from about 0.1 mil to about 0.4 mil; and the
30 core layer has such thickness as required to bring the polymer gauge thickness of the entire film to a thickness from about 1.5 mil to about 2.5 mil.

Although 3, 4, and 5 layer embodiments have been disclosed herein. Films of six or more layers are contemplated by the invention.

EXAMPLES

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Examples 1-3

Table 1 shows the compositions and optical properties of three exemplar films having five layers (L1 - L5). The top skin layer (L1) of each film incorporates a red or a blue tint.

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The data of Table 1 demonstrate that the omission from film 2 of the whitening agent (6% TiO₂) from the top tie layer (L2) and the cavitating agent (8% PBT) from the core layer (L3) increases the light transmission of the film from 24% to almost 100% clarity (film 3).

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Examples 4-5: Both films have the same 5-layer structure (L1 - L5), with added red/blue tints and no optical brightener added:

20 L1 Top skin layer (Propylene ethylene copolymer with added red and blue tints).
L2 Top tie layer (Polypropylene with 4% TiO₂).
L3 Core layer (Polypropylene with added cavitating agent).
L4 Bottom tie layer (Polypropylene).
L5 Bottom skin layer (Polypropylene).

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Films 4 and 5 each have a total polymer gauge thickness of 1.9 mil, whereas their optical gauge thickness (including voids) is 2.6 mil.

30 Table 2 lists the optical thickness (optical gauge) and polymer thickness (polymer gauge, also referred to as "poly gauge") of each of the exemplified films 1-5 in mil (thousandths of an inch). The optical gauge is the total thickness of the

film including voids. The polymer gauge is the residual thickness of the film without voids. The L2 gauge is the thickness of the top tie layer in mil.

The %LT value is the percent light transmittance through the film measured photometrically. L* is the lightness value; A* is the red/green rating; and B* is the blue/yellow rating according to the standard color measurement in the CIE system measured on a Hunterlab colorimeter. Opacity is the percent loss of light reflected through the film onto a white reflective tile surface and transmitted back through the film, measured photometrically.

Table 1

Examples 1-3

The following films are produced as 5-layer structures (L1-L5).
 Blue tint / red tints are incorporated into layer L1.

10	Film	L1	L2	L3	L4	L5	%LT
15	1.	PE + red/ blue tint	PP + 6% TiO ₂ + 0.2% OB	PP + cavitating agent	PP	PP + antiblocking additive	24
20	2.	PE + red/ blue tint	PP + 6% TiO ₂ + 0.2% OB	PP + cavitating agent	PP	PP + antiblocking additive	24
25	3.	PE + red/ Blue tint	PP + 0.2% OB	PP	PP	PP + antiblocking additive	99

PE: Propylene-ethylene copolymer.
 PP: Polypropylene.

Table 2

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Optical properties of the exemplified films

Film	Optical gauge Mil	Poly gauge mil	% LT	L*	A*	B*	Opacity [§] %
1	2.3	1.5	24	93.7	-0.61	-4.38	97.1
2	2.3	1.5	24	93.8	-0.71	-4.39	96.7
3	1.5	1.5	99	93.6	-0.51	-1.82	
4	2.6	1.9		93.2	-0.46	-4.78	96.3
5	2.6	1.9		93.8	-0.44	-4.22	96.8

[§] opacity measured against a white background.

WHAT IS CLAIMED IS:

1. A multilayer plastic film for imaging applications, comprising:
 - a) a core layer comprising polypropylene;
 - b) a top skin layer comprising a polyolefin polymer selected from the group consisting of: a propylene-ethylene copolymer, a high density polyethylene polymer, a low density polyethylene polymer, and polypropylene; and
 - c) a bottom skin layer comprising a polyolefin polymer selected from the group consisting of polypropylene, polyethylene and a polypropylene-ethylene copolymer; and
- 10 wherein at least one layer of the film includes at least one image enhancing colored tint.
2. The multilayer plastic film according to claim 1, wherein the image enhancing colored tint is a red tint selected from the group consisting of a quinacridone tint and a derivative of a quinacridone tint.
- 15 3. The multilayer plastic film according to claim 1, wherein the image enhancing colored tint is a blue pigment selected from the group consisting of a phthalocyanine blue pigment, a cromophthal blue pigment, an irgazin blue pigment, an irgalite organic blue pigment, a cobalt aluminate pigment, and a cobalt chromium aluminate inorganic blue pigment.
- 20 4. The multilayer plastic film according to claim 1, wherein said image enhancing colored tint is present in said top skin layer in an amount up to about 0.2% by weight.
5. The multilayer plastic film according to claim 4, further comprising a top tie layer positioned between the core layer and the top skin layer, wherein

said top tie layer comprises polypropylene, and wherein said top tie layer includes up to about 0.2% by weight of said image enhancing colored tint.

6. The multilayer plastic film according to claim 1, further comprising a top tie layer positioned between the core layer and the top skin layer, wherein
5 said top tie layer comprises polypropylene, and wherein said top tie layer includes up to about 0.2% by weight of said image enhancing colored tint.

7. The multilayer plastic film according to claim 6, further comprising a bottom tie layer positioned between the core layer and the bottom skin layer; said bottom tie layer comprising polypropylene.

10 8. The multilayer plastic film according to claim 6, wherein said top tie layer further comprises TiO_2 and an optical brightener.

9. The multilayer plastic film according to claim 6, wherein said core layer further comprises a cavitating agent, and said bottom skin layer further comprises an antiblocking agent.

15 10. The multilayer plastic film according to claim 6, wherein said top skin layer includes from about 0.005% to about 0.15% by weight of said image enhancing colored tint; and wherein said top tie layer includes from about 4% to about 6% by weight of TiO_2 and from about 0.05% to about 0.5% of an optical brightener; and wherein said core layer includes up to about 6%
20 by weight of a cavitating agent.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER		
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According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
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EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 441 694 A (USIPLAST) 14 August 1991 (1991-08-14) column 3, line 38 -column 4, line 40 claims 1,2,8; examples 1,2 ---	1-4
Y	WO 98 04403 A (MOBIL OIL CORP) 5 February 1998 (1998-02-05) page 2, line 4 - line 17 page 6, line 1 -page 7, line 5 examples 1-3 ---	1-10
Y	EP 0 578 255 A (NIPPON ZEON CO) 12 January 1994 (1994-01-12) column 7, line 27 - line 42 ---	1-10
A	WO 98 56662 A (MOBIL OIL CORP) 17 December 1998 (1998-12-17) page 4, line 31 -page 6, line 13 ---	1-10

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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